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OFFICE OF NAVAL RESEARCH

Grant N00014-90-J-1193

TECHNICAL REPORT No. 28

Surface-Induced Optical Bistability in Coupled Exciton-Phonon Systems

by

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Prepared for publication

in

Physics Letters A

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November 1990

UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

Form Approved OMB No. 0704-0188

1a. REPORT SECURITY CLASSIFICATION: Unclassified

1b. RESTRICTIVE MARKINGS

2a. SECURITY CLASSIFICATION AUTHORITY

2b. DECLASSIFICATION / DOWNGRADING SCHEDULE

3. DISTRIBUTION / AVAILABILITY OF REPORT: Approved for public release; distribution unlimited

4. PERFORMING ORGANIZATION REPORT NUMBER(S): UBUFFALO/DC/90/TR-28

5. MONITORING ORGANIZATION REPORT NUMBER(S)

6a. NAME OF PERFORMING ORGANIZATION: Depts. Chemistry & Physics, State University of New York

6b. OFFICE SYMBOL (If applicable)

6c. ADDRESS (City, State, and ZIP Code): Fronczak Hall, Amherst Campus, Buffalo, New York 14260

7a. NAME OF MONITORING ORGANIZATION

7b. ADDRESS (City, State, and ZIP Code): Chemistry Program, 800 N. Quincy Street, Arlington, Virginia 22217

8a. NAME OF FUNDING / SPONSORING ORGANIZATION: Office of Naval Research

8b. OFFICE SYMBOL (If applicable)

9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER: Grant N00014-90-J-1193

8c. ADDRESS (City, State, and ZIP Code): Chemistry Program, 800 N. Quincy Street, Arlington, Virginia 22217

10. SOURCE OF FUNDING NUMBERS

PROGRAM ELEMENT NO.	PROJECT NO.	TASK NO	WORK UNIT ACCESSION NO.

11. TITLE (Include Security Classification): Surface-Induced Optical Bistability in Coupled Exciton-Phonon Systems

12. PERSONAL AUTHOR(S): D. L. Lin, Xiao-shen Li and Thomas F. George

13a. TYPE OF REPORT

13b. TIME COVERED FROM ______ TO ______

14. DATE OF REPORT (Year, Month, Day): November, 1990

15. PAGE COUNT: 16

16. SUPPLEMENTARY NOTATION: Prepared for publication in *Physics Letters A*

17. COSATI CODES

FIELD	GROUP	SUB-GROUP

18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)

OPTICAL BISTABILITY, SURFACE-INDUCED, EXCITON-PHONON SYSTEM, REFLECTING SURFACE, POLYDIACETYLENE, THEORY

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

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20. DISTRIBUTION / AVAILABILITY OF ABSTRACT: ☒ UNCLASSIFIED/UNLIMITED ☒ SAME AS RPT ☐ DTIC USERS

21. ABSTRACT SECURITY CLASSIFICATION: Unclassified

22a. NAME OF RESPONSIBLE INDIVIDUAL: Dr. David L. Nelson

22b. TELEPHONE (Include Area Code): (202) 696-4410

22c. OFFICE SYMBOL

Physics Letters A, in press

Surface-induced optical bistability in coupled exciton-phonon systems

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Abstract

Nonlinear optical responses of exciton-phonon-coupling systems near a reflecting surface are investigated. For a polydiacetylene chain lying parallel to the surface, optical bistability induced by the surface is discovered. It is also found that the threshold and contrast of the bistability can be controlled by adjusting the distance of the system from the surface, and that vacuum fluctuations are reduced whenever the bistability occurs. The nature of the bistability and its origin are discussed.

1990 PACS Numbers: 42.65.-k, 42.65.pc, 36.20.-r, 78.65.Hc

Ever since the mid-1940's, it has been known[1] that the spontaneous emission probability of a spin system can be dramatically influenced by its environment. In recent years, inhibited spontaneous emission has been investigated extensively in atomic systems[2] and in solids.[3] It is found that the spontaneous radiation rate is altered markedly by merely changing the surrounding material with a different index of refraction. On the other hand, a great deal of work has been devoted to the study of an adatom near a solid surface.[4,5] Optical properties are radically changed because of the presence of the surface, and the change depends on the structure of the substate material. Such problems are of fundamental importance as well as of practical interest. Fundamentally, one has to develop a quantum mechanical theory for dissipative systems to deal with such nonlinear interactions between the atom and the substrate which does not possess bulk symmetry. From a practical point of view, such studies can lead to applications in surface photochemistry and surface optics.

Instead of atoms or molecules adsorbed at a solid surface, we consider an exciton-phonon coupling system such as a one-dimensional semiconductor of polymers. In particular, we consider a sample of polydiacetylene (PDA) located near a metal surface. The system closely resembles the experimental situation[6] in which PDA films are sandwiched between other materials. PDA is a good candidate for such considerations because of its large third-order susceptibility $\chi^{(3)}$,[7] and because of its small transmission loss α.[8] Thus it possesses a fairly large ratio $\chi^{(3)}/\alpha$, which is usually a measure of the usefulness of a material employed in switching devices. Moreover, PDA is more flexible to use in the construction of waveguides.[9]

For a linear chain of PDA-toluene-sulfonate (PTS) placed at a distance d from and oriented parallel to the metal surface, we shall study its response

to a laser beam shining on it. Novel phenomena have been found recently in the study of the nonlinear optical response of PTS in a thin film in the transient regime,[10] and in an optical cavity in the steady-state regime.[11] In this Letter, our purpose is to investigate the possibility of optical multistability induced by the metal surface, even though there is no cavity to feedback the incident signal. The light-induced excitons in PTS act as emitting dipoles. The emitted light is reflected by the metal surface and interacts with the excitons themselves, and changes their dynamical behavior. In addition, the excitons are also coupled to phonon modes in PTS.[12,13] Such coupling plays a very important role in the nonlinear optical response of polymers.[10-12]

This complicated interacting system is treated by modeling the excitons and phonon modes by damped oscillators and by assuming dissipative interactions[14] between the exciton and the reflected field. Thus the problem is described by the nonhermitian Hamiltonian

$$H = H_x + H_p + H_{xp} + H_{xf} + H_{xR} \quad , \tag{1}$$

where the free exciton energy operator is

$$H_x = \hbar(\omega_x - i\frac{\gamma_x}{2})\, a^\dagger a \quad , \tag{2}$$

the free phonon energy is

$$H_p = \hbar \sum_i (\omega_i - i\frac{\gamma_i}{2})\, b_i^\dagger b_i \quad , \tag{3}$$

the exciton-phonon interaction energy is

$$H_{xp} = \hbar \sum_i \lambda_i \; a^\dagger a(b_i + b_i^\dagger) \quad , \tag{4}$$

the interaction energy between the exciton and driving field in the rotating-wave approximation (RWA) is

$$H_{xf} = -(\mu a E_0^* e^{i\omega_0 t} + \mu^* a^\dagger E_0 e^{-i\omega_0 t}) \quad , \tag{5}$$

and the interaction energy between the exciton and surface-reflected field in the RWA is

$$H_{xR} = -\mu^* a^\dagger E_R \quad . \tag{6}$$

The notation is as follows. The operators $a^\dagger(a)$ and $b_i^\dagger(b_i)$ create (annihilate) an exciton with energy $\hbar\omega_x$ and a phonon in the i-th mode with energy $\hbar\omega_i$, respectively. The corresponding decay rates are denoted by γ_x and γ_i, respectively, and λ_i is a coupling constant. The dipole moment is given as $p = \mu a$ with μ as its matrix element, and the driving field is $E = E_0 \exp(i\omega_0 t)$ with amplitude E_0 and frequency ω_0. E_R stands for the reflected field at the position of the dipole. It is proportional to p and hence can be written as $E_R = E_r a$, where E_r is a c-number. As we shall see later, E_R is in general a complex quantity, so that the interaction (6) is dissipative. We also note that the momentum dependence of the exciton has been neglected in this Hamiltonian.[10-12]

Following Dekker's quantization procedure for dissipative systems,[15] the equation of motion for the density operator of the PTS absorbate can be written as

$$\dot{\rho} = -\frac{i}{\hbar}[a^\dagger,[a,H]\rho] + \frac{i}{\hbar}[\rho[H^\dagger,a^\dagger],a]$$

$$-\frac{i}{\hbar}\sum_i [b_i^\dagger,[b_i,H]\rho] + \frac{i}{\hbar}\sum_i [\rho[H^\dagger,b_i^\dagger],b_i] \quad . \tag{7}$$

Since we are interested in the scattered field intensity I_{sc} as a function of the input optical field intensity I_{in}, the effects of quantum fluctuations are negligible,[16] and a semiclassical approach is applicable. Thus it is sufficient to use the mean values $\alpha = \langle a \rangle$, $n = \langle a^\dagger a \rangle$ and $\beta_i = \langle b_i \rangle$. The equation of motion for these variables in the Schrödinger picture can be obtained directly from Eq. (7). In the rotating frame, we have

$$\dot{\alpha} = \langle \dot{a} \rangle = \mathrm{Tr}(\dot{\rho} a)$$

$$= -[i(\Delta+\omega_s) + \frac{1}{2}\gamma_x + \gamma_s]\alpha + i\Omega - i\sum_i \lambda_i(\beta_i+\beta_i^*)\alpha \tag{8}$$

$$\dot{n} = -(\gamma_x+2\gamma_s)n - i\Omega^*\alpha + i\Omega\alpha^* \tag{9}$$

$$\dot{\beta}_i = -(i\omega_i+\frac{1}{2}\gamma_i)\beta_i - i\lambda_i n \quad , \tag{10}$$

where we have defined the detuning $\Delta = \omega_x - \omega_0$ and the Rabi frequency $\Omega = \mu^* E_0$. The surface effect on the exciton is reflected in the surface-induced frequency shift

$$\omega_s = |\mu|^2 \mathrm{Re}(E_r/\mu) \tag{11a}$$

and the decay rate

$$\gamma_s = |\mu|^2 \mathrm{Im}(E_r/\mu) \quad . \tag{11b}$$

The reflected electric field E_r in the dipole direction can be found by a classical approach. We take the oscillating dipole moment to be located at a distance d from the surface of a semi-infinite metal. From electromagnetic field theory, we find in a straightforward manner that

$$E_r^{\parallel} = \frac{i}{2} \sqrt{\epsilon}\ k_0^3\ \mu \int_0^{\infty} du\ u[R_{\perp} + (1-u^2)R_{\parallel}]/\sqrt{1-u^2} \tag{12}$$

when the dipole $\vec{p}$ is parallel to the surface, and

$$E_r^{\perp} = -i\sqrt{\epsilon}\ k_0^3\ \mu \int_0^{\infty} du\ u^3 R_{\parallel}/\sqrt{1-u^2} \tag{13}$$

when the dipole is perpendicular to the surface. Here we have defined $k_0 = \omega/c$ and

$$R_{\parallel} = \frac{\epsilon_1\mu_2 - \epsilon_2\mu_1}{\epsilon_1\mu_2 + \epsilon_2\mu_1} \quad , \tag{14a}$$

$$R_{\perp} = \frac{\mu_1 - \mu_2}{\mu_1 + \mu_2} \tag{14b}$$

$$\mu_{\ell} = \sqrt{\epsilon_{\ell}/\epsilon_1 - u^2} \quad , \qquad \ell = 1,2 \tag{14c}$$

with ϵ as the dielectric constant and the subscripts 1 and 2 labelling the medium containing the PTS chain and the metal substrate, respectively. For definiteness, we consider the case where $\vec{p}$ is parallel to the surface. It then follows directly from Eqs. (11) and (12) that

$$\gamma_s = \frac{3}{4}\gamma_0 \ \mathrm{Im}\int_0^\infty du\ u[(1-u^2)R_\parallel + R_\perp]e^{i\mu_1 x}/\mu_1 \quad , \tag{15a}$$

$$\omega_s = -\frac{3}{4}\gamma_0 \ \mathrm{Re}\int_0^\infty du\ u[(1-u^2)R_\parallel + R_\perp]e^{i\mu_1 x}/\mu_1 \quad , \tag{15b}$$

where

$$\gamma_0 = \frac{2}{3}\sqrt{\epsilon_1}\left(\frac{\omega_x}{c}\right)^3 |\mu|^2 = \frac{1}{2}\gamma_x \tag{16}$$

is the decay rate in the absence of the surface,[17] and $x = 2d\sqrt{\epsilon_1}\omega_x/c$.

In the steady state, the solution for α can easily be found from Eqs. (8)-(10) as

$$\alpha = \Omega/[\Delta+\omega_s-\lambda_p n-i(\gamma_s+\tfrac{1}{2}\gamma_x)] \quad , \tag{17}$$

where λ_p is defined by

$$\lambda_p = 2\sum_i \lambda_i^2\omega_i/(\omega_i^2+\tfrac{1}{4}\gamma_i^2) \simeq 2\sum_i \lambda_i^2/\omega_i \tag{18}$$

because $\omega_i \gg \gamma_i$. The mean number of excitons is determined by the equation

$$\lambda_p^2 n^3 - 2(\Delta+\omega_s)\lambda_p n^2 + [(\Delta+\omega_s)^2 + (\gamma_s+\tfrac{1}{2}\gamma_x)^2]n - I_{in} = 0 \quad , \tag{19}$$

where the input laser intensity is $I_{in} = |\Omega|^2$. In the scattering region where the incident field vanishes, it is known that the positive-frequency part of the field may be written as[18]

$$E^{(+)}(\vec{r},t) = E_f^{(+)}(\vec{r},t) - \frac{\omega_x^2}{4\pi c^2 r^3} (\vec{p} \times \vec{r}) \times \vec{r}\, a(t-\frac{r}{c}) \quad , \tag{20}$$

where $E_f(\vec{r},t)$ describes the free propagation. The mean value of the scattered intensity, in the steady state is thus given by

$$\begin{aligned} I_{sc} &= I(\vec{r})/I_0(\vec{r}) = |\alpha|^2 \\ &= I_{in}/\{[\Delta+\omega_s-\lambda_p n(I_{in})]^2 + (\gamma_s+\tfrac{1}{2}\gamma_x)^2\} \quad , \end{aligned} \tag{21}$$

where

$$I_0(\vec{r}) = [\frac{\omega_x^2}{4\pi c^2 r^3} (\vec{p} \times \vec{r}) \times \vec{r}]^2 \quad . \tag{22}$$

The intensity I_{sc} is studied numerically as a function of I_{in} according to Eq. (21) for a PTS chain parallel to the surface of an ideal metal for which $\epsilon_2 \to \infty$. As a realistic approximation, we have assumed that only four phonon modes[13] are strongly coupled to excitons in PTS. The parameters for these modes are:[10-13] $\omega_1 = 5.16$, $\lambda_1 = 2.0$; $\omega_2 = 3.68$, $\lambda_2 = 1.66$; $\omega_3 = 2.98$, $\lambda_3 = 0.46$; and $\omega_4 = 2.36$, $\lambda_4 = 0.48$. For simplicity we have chosen $\frac{1}{2}\gamma_x$ to be

the unit for all quantities of the dimension t^{-1} in our calculations. Optical bistability is found when the input field frequency matches the exciton frequency. Some of our results for the resonant excitation ($\Delta = 0$) are plotted in Fig. 1, clearly demonstrating that the bistability is induced by the surface. Our numerical calculation shows a strong dependence of the optical bistability on the distance d of PTS from the surface, especially when $x \leq 2.5$. As x decreases further, it is observed that the contrast of the bistability (the ratio of the intensity of the upper branch to that of the lower branch) increases markedly, and that the threshold (the smallest input field intensity for multiple solutions of I_{sc}) decreases at the same time. Furthermore, we have also found that the optical bistability appears only when γ_s becomes negative. In other words, reduced vacuum fluctuations always accompany the optical bistability.

It has been established both experimentally[2,19] and theoretically[17] that the spontaneous emission changes dramatically when the atomic system is near a reflecting surface or between two mirrors. This implies that the mode structure of the vacuum field suffers significant modification because of the finite geometry. A similar situation in solid-state physics and electronics has also been reported, and the spontaneous electron-hole recombination rate can be controlled by adjusting the mirror position or by varying the index of refraction of the medium.[3] What we have found here is that by adjusting the environmental conditions to reduce the radiation rate, one can not only produce low-noise lasers from a semiconductor quantum well but also create optical bistability from a polymer chain near a reflecting surface.

The origin of the optical bistability may be understood by looking at the driving frequency dependence of the scattered field intensity. For a

fixed driving field intensity, its frequency can be scanned over the near-resonance region. Thus the exciton number becomes a function of ω_0, as can be seen from Eq. (19). Consequently, I_{sc} can be regarded as a function of ω_0 also. Figure 2 shows the surface-induced optical bistability from the point of view of the driving field frequency instead of its intensity. It is found that there are three possible absorption rates for a certain small frequency range. The scattered intensity in the higher branch corresponds to the higher absorption rate, and in the lower branch to the lower absorption rate. Since the absorption part of the optical susceptibility is proportional to the scattered field intensity for fixed input intensity and distance x, the bistability described above is mainly an absorptive one. In other words, the surface-induced absorption rate is responsible for this particular optical bistability. This is easily understood from energy considerations, because a higher output intensity of the excitonic dipole results from more effective absorption of the input energy, and vice versa.

Acknowledgment

This work was supported in part by the Office of Naval Research.

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Figure Captions

1. Scattered field intensity I_{sc} vs the driving field intensity I_{in} for various distances x when $\Delta = 0$: (a) $x = \infty$, (b) $x = 1.5$, (c) $x = 1.1$, (d) $x = 0.9$ and (e) $x = 0.85$.

2. I_{sc} as a function of the driving field frequency ω_0 for a fixed $I_{in} = 0.10$: (a) $x = \infty$ and (b) $x = 1.5$.

Fig. 1

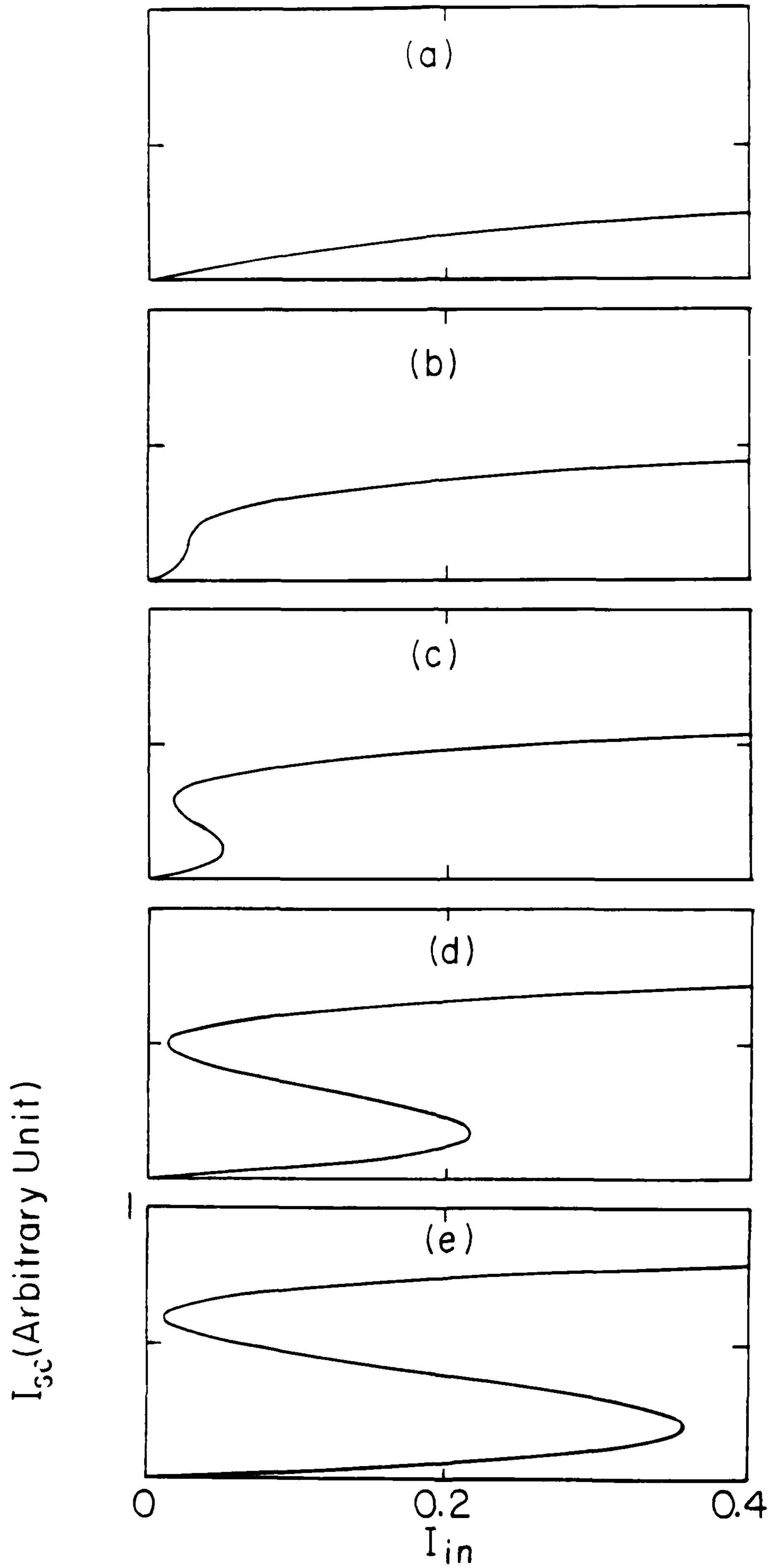
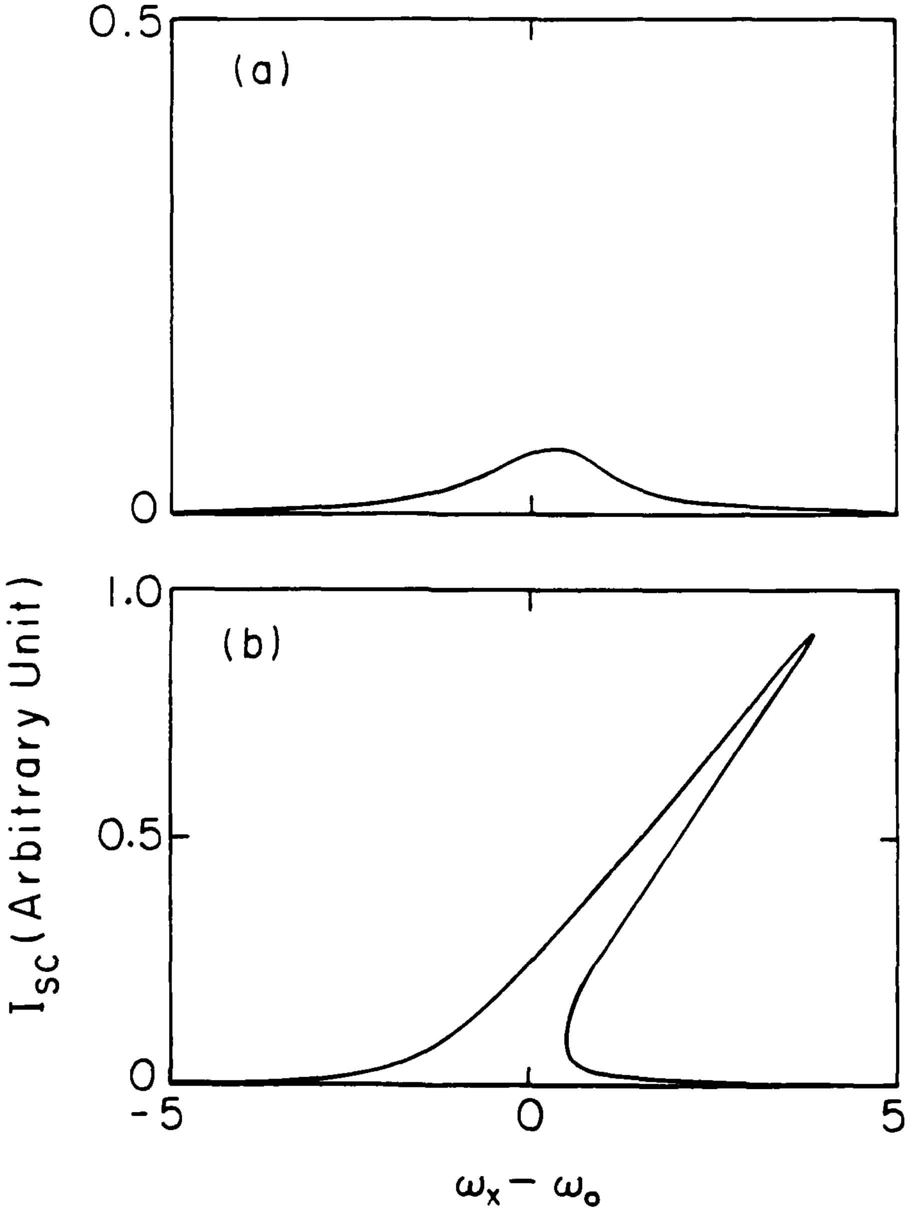


Fig. 2



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